AD-A133 864 HIGH TEMPERATURE CATALYTICALLY ASSISTED COMBUSTION(U) 1/1
PRINCETON UNIV NJ DEPT OF MECHANICAL AND AEROSPACE
ENGINEERING F V BRACCO ET AL. 28 JAN 83
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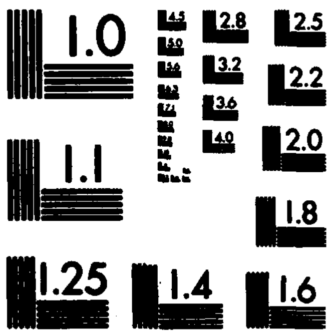

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Catalytic Combustion

20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

Results of research on a two-dimensional, transient catalytic combustion model and on a high-temperature perovskite catalyst are presented. A recently developed two-dimensional, transient model has been used to study the ignition of CO/air mixtures in a platinum coated catalytic honeycomb. Comparisons between calculated and measured steady state substrate temperature profiles and exhaust gas compositions show good agreement. A platinum-doped perovskite catalyst has been designed to exhibit low temperature light off and (over)

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high temperature stability. Preliminary tests using a perovskite powder with one percent by weight platinum are encouraging, showing very little change in surface activity when used with propane fuel.

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HIGH TEMPERATURE CATALYTICALLY ASSISTED COMBUSTION

AFOSR-TR- 83-0866

STATUS

The major accomplishments during the past year have been the development and testing a two-dimensional transient catalytic combustion model and the design and testing a new catalyst material with low temperature light-off and high temperature stability characteristics. The accomplishments in these two areas are summarized in the following paragraphs.

A recently developed two-dimensional transient model has been used to study the ignition of CO/air mixtures in a platinum coated catalytic honeycomb. This model includes coupling between the gas and substrate, radiative heat loss to the outside and radial heat losses providing solutions for the two-dimensional temperature field in the substrate and the two-dimensional temperature, composition and velocity field in the gas. The model has been used to predict the transient and steady state using inlet conditions for which steady state experimental data is available. In the calculation the inlet conditions for velocity and temperature are set and the transient begins by injecting fuel. This model is particularly useful for predicting the temperature gradients in the substrate which in turn can be used to predict the thermal stresses in the catalyst.

An example of the transient behavior of the substrate axial temperature profile in response to a 100 millisecond fuel ramp is shown in Figure 1. It can be seen that during the early part of the transient there are very large spatial and temporal gradients near the entrance. The model also shows that the heat release producing these gradients occurs primarily at the entrance is due to heterogeneous reactions and is diffusion controlled. This example demonstrates how the model can be used to gain insight into the relative importance of the numerous physical and chemical processes which control the overall catalyst performance. Since transient measurements are not available the calculated steady state results are compared to steady state measurements. The predicted steady state catalyst temperature profile and the exhaust gas composition are found to agree well with measurements over a range of inlet velocities and equivalence ratios. A comparison of predicted (solid line) and measured (symbols) substrate temperature profiles is shown in Figure 2.

Operating temperatures of catalytic combustors often exceed 1500 K, resulting in unacceptably short lifetimes for standard catalysts. A new

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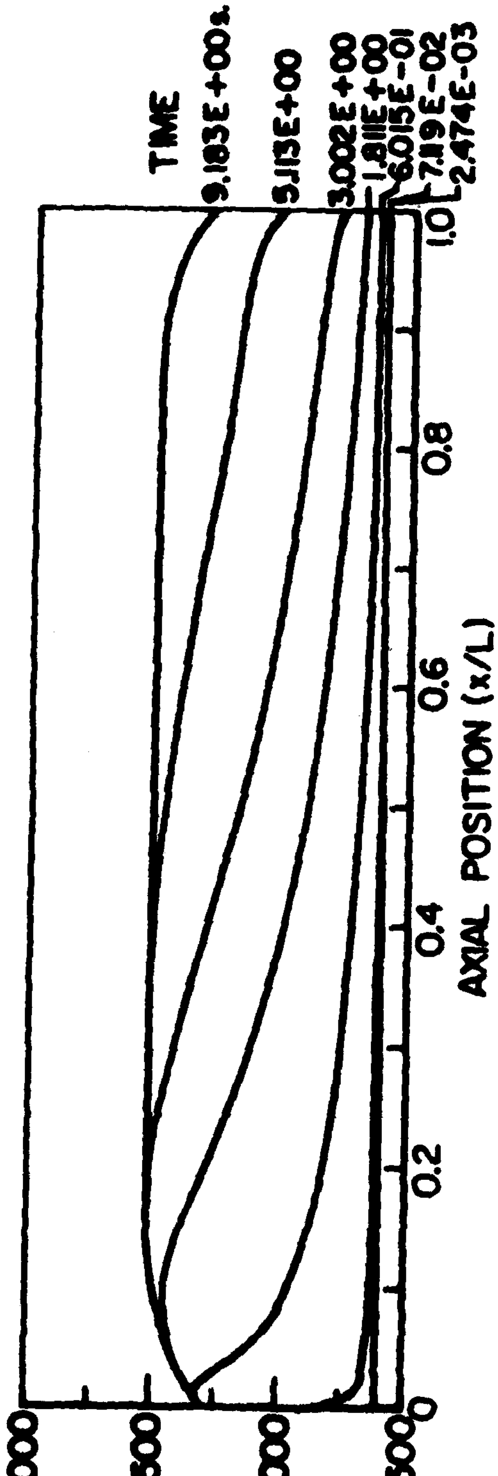

ure 1: Transient behavior of the substrate axial temperature profile in response to a 100 millisecond fuel ramp.

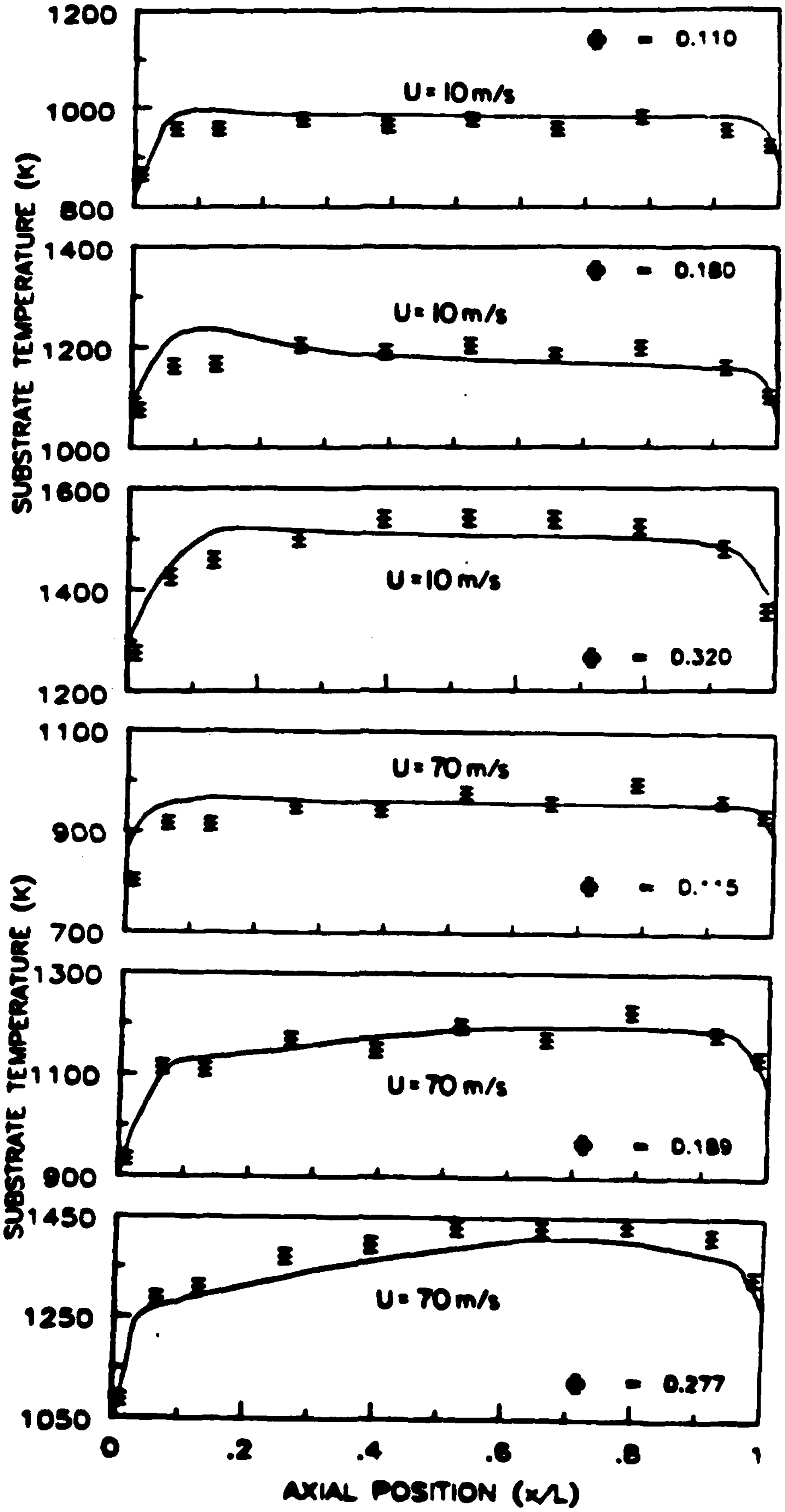

Figure 2: A comparison of predicted (solid line) and measured (symbols) steady state substrate temperature profiles.

catalyst has been designed to exhibit long lifetime at high temperatures and adequate ignition characteristics at low temperatures. The catalyst is a modified perovskite based on La $(Cr_{0.5}\ Al_{0.5})\ O_3$. This ceramic has been doped to make it electrically conductive and consequently it can be resistively heated to bring the catalyst up to the required light-off temperature. In addition, platinum has been incorporated into the crystal structure to give improved low temperature light-off while having a low platinum vapor pressure at high operating temperatures. This catalyst can be used in powdered form, by washcoating it onto a high temperature ceramic substrate, or as sintered monolythic structures, e.g. plates. The advantage of using the catalyst in the form of plates is that they can be resistively heated to assist light-off. However, the catalyst is more readily available in powdered form and therefore the first tests with the new catalyst have been made with it washcoated on a honeycomb substrate. The substrate used was mullite, three inches long with 1/16 inch square cells. Experiments using a pure platinum washcoat and a perovskite powder with nominal 1.0% (by weight) platinum (supplied by Prof. Harlan Anderson, University of Missouri-Rolla) washcoat have been made. The tests consist of establishing the light-off temperature and low temperature performance for several equivalence ratios and inlet velocities, after which the catalyst is "aged" for several hours at 1500 °K and then the light-off and low temperature experiments repeated. The purpose of running at low temperature is to insure that all of the heat release is due to surface reactions. Therefore the maximum substrate temperature in these runs was kept below 800°C. Even at low temperatures, however it is important that the overall process be surface reaction rate controlled and not diffusion controlled. If the heat release is surface reaction rate controlled, then the measured substrate temperature profiles will provide an indication of changes in surface activity. Figure 3 shows measured substrate temperature profiles for the perovskite with nominal 1% by weight platinum using propane fuel at an equivalence ratio of 0.3, inlet temperature of 698°K, inlet velocity of 6 m/sec, and 1 atmosphere pressure. The before aging result is with the new catalyst. Also shown are results after four successive aging runs. The indicated times are cumulative. As the results show, there is a noticeable loss of activity after the second and fourth aging runs which were done with a mixture of hydrogen and propane. However, after the first

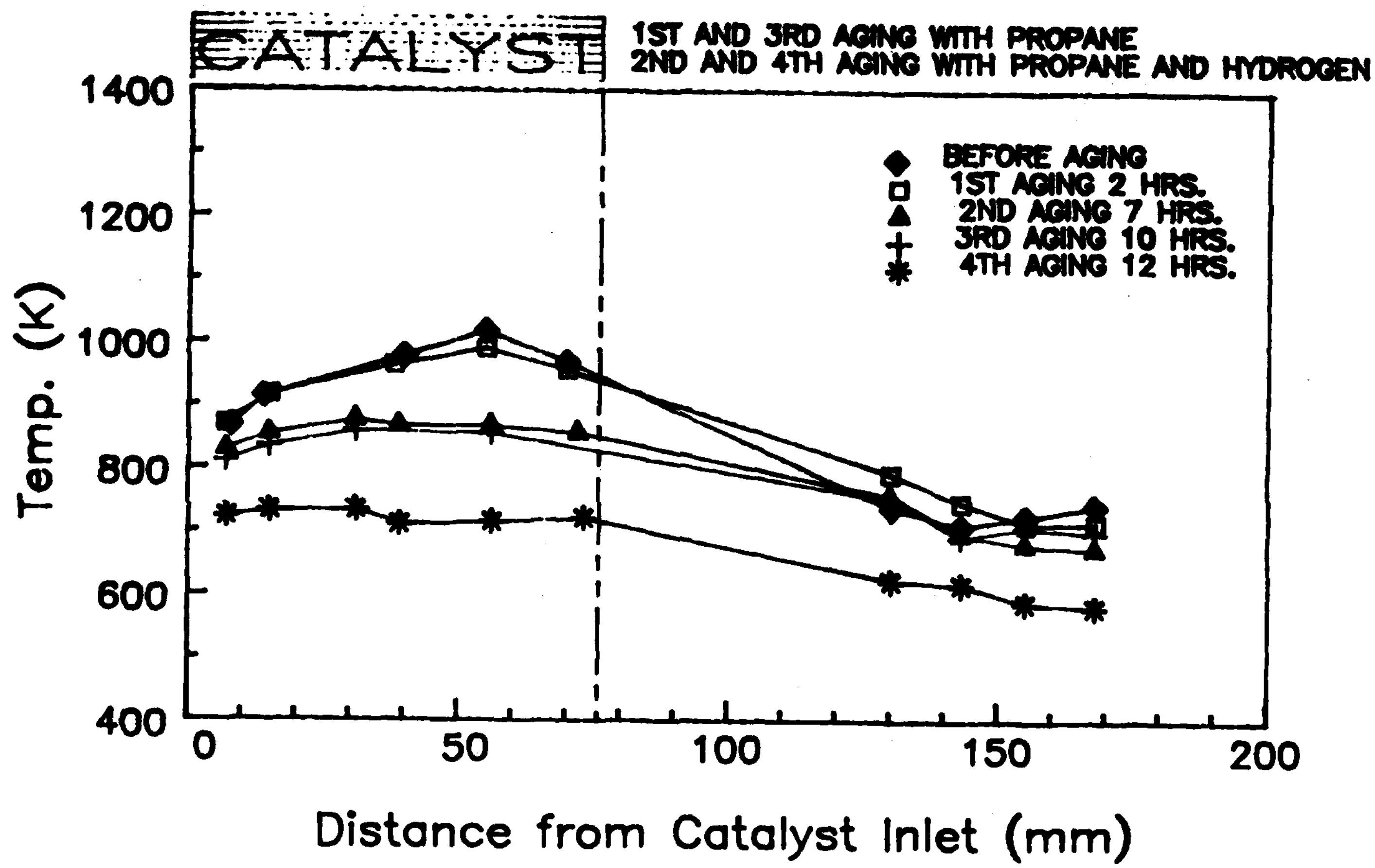

Figure 3: Perovskite with nominal 1% by weight
Platinum catalyst
Propane fuel, equivalence ratio = 0.30
Inlet temperature = 698°K, inlet velocity = 6.0 m/s

and third aging runs, which were done with pure propane, there is little if any indication of reduced surface activity. These results are very encouraging, indicating very little change in surface activity when used with propane fuel. The tests with hydrogen however indicate a type of poisoning effect which quickly reduces the activity of the catalyst. Results from these tests will be used as a guide in the selection of the optimum platinum content, in terms of both adequate low temperature light-off and minimal high temperature aging, to be used in the catalytic plates which will be produced and tested in the near future.

PUBLICATIONS

1. Bruno, C., Walsh, P.M., Santavicca, D.A., Sinha, N., Yaw, Y. and Bracco, F.V., "Catalytic Combustion of Propane/Air Mixtures on Platinum". Accepted for publication in Combustion Science and Technology.

2. Bruno, C., Walsh, P.M., Santavicca, D.A. and Bracco, F.V., "High Temperature Catalytic Combustion of $CO/O_2/N_2$, Ar, He, CO_2/H_2O Mixtures on Platinum." Accepted for publication in the International Journal of Heat and Mass Transfer.

3. Sinha, N., Bruno, C. and Bracco, F.V., "Two-Dimensional, Transient Catalytic Combution of CO/Air on Platinum". Submitted to the International Journal of Heat and Mass Transfer.

PERSONNEL

The following professional personnel have contributed to this research.

Professor F.V. Bracco, Associate Professor

Professor B.S.H. Royce, Professor

Dr. D.A. Santavicca, Research Engineer

Dr. C. Bruno, Research Staff Member

Dr. P. Curtis, Research Staff Member

Mr. N. Sinha, Graduate Student - MSE degree, August 1982

Ms. Y. Stein, Technical Staff Member

INTERACTIONS

1. The following paper was presented at the Eastern States Section of the Eastern States Section of the Combution Institute Fall Meeting, December 1982.

   "Perovskite Catalysts for High Temperature Catalytically Assisted Combustion" by D.A. Santavicca, Y. Stein and B.S.H. Royce.

2. A presentation on the catalytic combustion of propane was given at the 5th EPA Workshop on Catalytic Combution, San Antonio Texas, September 1981.

3. The following people have visited our laboratory during the past year for technical discussions on catalytic combustion.

   Dr. L.L. Hegedus, W.R. Grace and Company
   Dr. J. Latty, Dresser Industries

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